Mangalore University

Synopsis of the **Pre-PhD thesis submission colloquium**

on

Trace elements and boron isotopic study in marine carbonates: Reconstruction of paleo-pH and oceanographic conditions

for the award of the

Doctor of Philosophy

in

Marine Geology (under the Faculty of Science)

by

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September 2021

Mangalore University

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Trace elements and boron isotopic study in marine carbonates: Reconstruction of paleo-pH and oceanographic conditions

Introduction

Over geologic timescale (millions of years) planet Earth is maintained habitable by the CO_2 concentrations of the atmosphere which reflects a constant interplay between supply through mantle degassing and consumption by chemical weathering of silicate rocks (Sigman et al., 2010). In the short term (thousands of years), earth's orbital parameters, collectively known as Milankovitch Cycles controls the earth's climate by the partitioning of CO_2 between the atmosphere and the oceans. Internal feedbacks like planetary albedo, winds, ocean circulation and biological productivity also play an important role in modulating the climate. Overall, the climate of the past has rarely remained invariant and Planet Earth has experienced a large natural variation, covering a wide range of amplitude and frequency of its climate throughout its history. These natural climate shifts and perturbations provide valuable information on the functioning and sensitivity of the climate system (Mackenzie and Andersson, 2013). However, these past climatic variations are rarely directly recorded. Therefore, we have to rely on proxies for past climate reconstruction. The direct records, such as the Antarctic Ice core based pCO_2 history is derived from the air bubbles trapped in the Ice core and allows us to reconstruct the earth's atmospheric composition going back to few hundred thousand years. The atmospheric partial pressure of CO_2 (pCO_2) measured in air bubbles from the ice cores fluctuates approximately between 180 ppm and 280 ppm during the quaternary glacial and interglacial periods respectively (Luthi et al., 2008). Recently, CO_2 concentration in the atmosphere has increased from ca. 280 ppm before preindustrial time (~1850) to ca. 415 ppm at present. Now, it is well established that the release of greenhouse gases (principally CO_2) due to the burning of fossil fuels is responsible for the temperature rise of ~0.6 - 0.8 °C since the industrial revolution (IPCC, 2021). About 20–30% of the anthropogenic CO_2 ejected in atmosphere during the last century has been absorbed by the ocean. This absorption has resulted a decrease in the surface ocean pH by approximately

0.1 units (from 8.2 to 8.1). This process of decrease in ocean pH is called ocean acidification (OA) (Doney et al., 2009).

Ocean acidification combined with global warming poses a serious threat to calcifying marine organisms (Hoegh-Guldberg et al., 2007). The impacts of OA on marine organisms and marine ecosystems have far-reaching consequences that will affect billions of people depending on the ocean for their food availability and livelihoods (Doney et al., 2020). Considering the importance of CO_2 and pH in the planning and mitigation strategies, it is important to have more records of CO_2 and pH and better understand the mechanism of pH and pCO_2 variability. Since the direct record of pCO_2 from the ice core is available only for the time span of past 800 kilo years. One way to look at climate history and transitions in the past is by using various proxies (Rae et al., 2021). Boron isotope ($\delta^{11}B$) in marine biogenic carbonates such as coral and foraminifera is used as a proxy to reconstruct the past pCO_2 and pH of the ocean. In the present thesis, boron isotopic composition ($\delta^{11}B$) and trace-element/Ca ratio of the corals and foraminifera from the Arabian Sea has been used to reconstruct the past pH/pCO_2 and paleo-oceanographic conditions of the Arabian Sea and effort has been made to understand the factors/forcing responsible for pH and pCO_2 variability.

Objectives of the research

Specific scientific objectives proposed for the thesis are as follows

- To reconstruct annual scale pH evolution history of the Arabian Sea from the boron isotopic study of corals.
- To understand the factors responsible for pH variability on annual to decadal scale and estimate the relative contribution of the different forcing factors to the overall pH variability.
- To reconstruct the pH evolution history of the Arabian Sea during the late Quaternary based on boron isotope ratio in foraminifera extracted from sediment cores.

- To reconstruction the paleoceanographic and paleoenvironmental condition of the study area from the trace metals/calcium ratios.

Outline of the thesis

In order to achieve these objectives, this thesis has been structured in the form of six chapters. In each chapter, except Chapter 1 (Introduction), an effort has been made to expound the proposed thesis objectives. To summarize, this thesis provides an important contribution to the understanding of forcing factors and mechanisms for pH and pCO_2 variability both at sub-annual to decadal and multi-millennial scales. Corals are an excellent archive to reconstruct the high-resolution climate history comparable to the instrumental records. In this study, we used the corals from the Arabian Sea to reconstruct the records of ocean pH from 1990 to 2013 at sub-annual resolution. Further, we have determined the recent ocean acidification trend in the Arabian Sea and investigate various factors contributing to pH variability and trend. This study utilizes planktic foraminifera $\delta^{11}B$ derived pH and pCO_2 record provides insight into the CO_2 source and sink variability of the Arabian Sea at glacial-interglacial scales. A brief overview of each chapter of the thesis has been presented below.

Chapter 1

Introduction

This Chapter describes the importance and significance of the proposed work, based on available literature on boron isotope and trace-metal/Ca ratio as a proxy for past ocean pH, pCO_2 and paleoceanographic conditions. A brief overview of climate during the geologic past and recent climate change due to anthropogenic CO_2 rise and its effects on marine organisms has been provided. Different proxy-based reconstructions of past pCO_2 along with their advantages and limitations/uncertainties have been briefly discussed followed by a detailed description of boron

isotope pH/pCO_2 proxy along with its caveats, merits and demerits over other proxies. A detailed description of the chemistry of ocean acidification and its effects on marine calcifiers has also been provided. This chapter also delineates the key knowledge gaps in the application of boron pH-pCO_2 proxy. Based on these key knowledge gaps, the chapter concludes with some key questions that are set as the objectives of this doctoral thesis.

Chapter 2

Materials and Methods

This chapter describes the importance of the study area for paleoclimate and paleoceanographic reconstructions and discusses the different techniques of sample preparation, processing, and analysis. In this thesis, two types of samples viz. coral and planktic foraminifera were used to reconstruct the pH/pCO_2 at sub-annual to decadal and millennial-scale respectively. The location of these samples has been shown in Figure 1. Trace-element/calcium ratios and boron isotopic composition of the biocarbonates (Foraminifera and corals) used in this study were determined using HR-ICPMS and MC-ICPMS respectively. Trace-element and $\delta^{11}B$ measurements in biogenic carbonates have been always challenging tasks due to matrix effect, mass biasing, polyatomic and organic interferences and large mass fractionation during analysis and sample preparation (Misra et al., 2014).

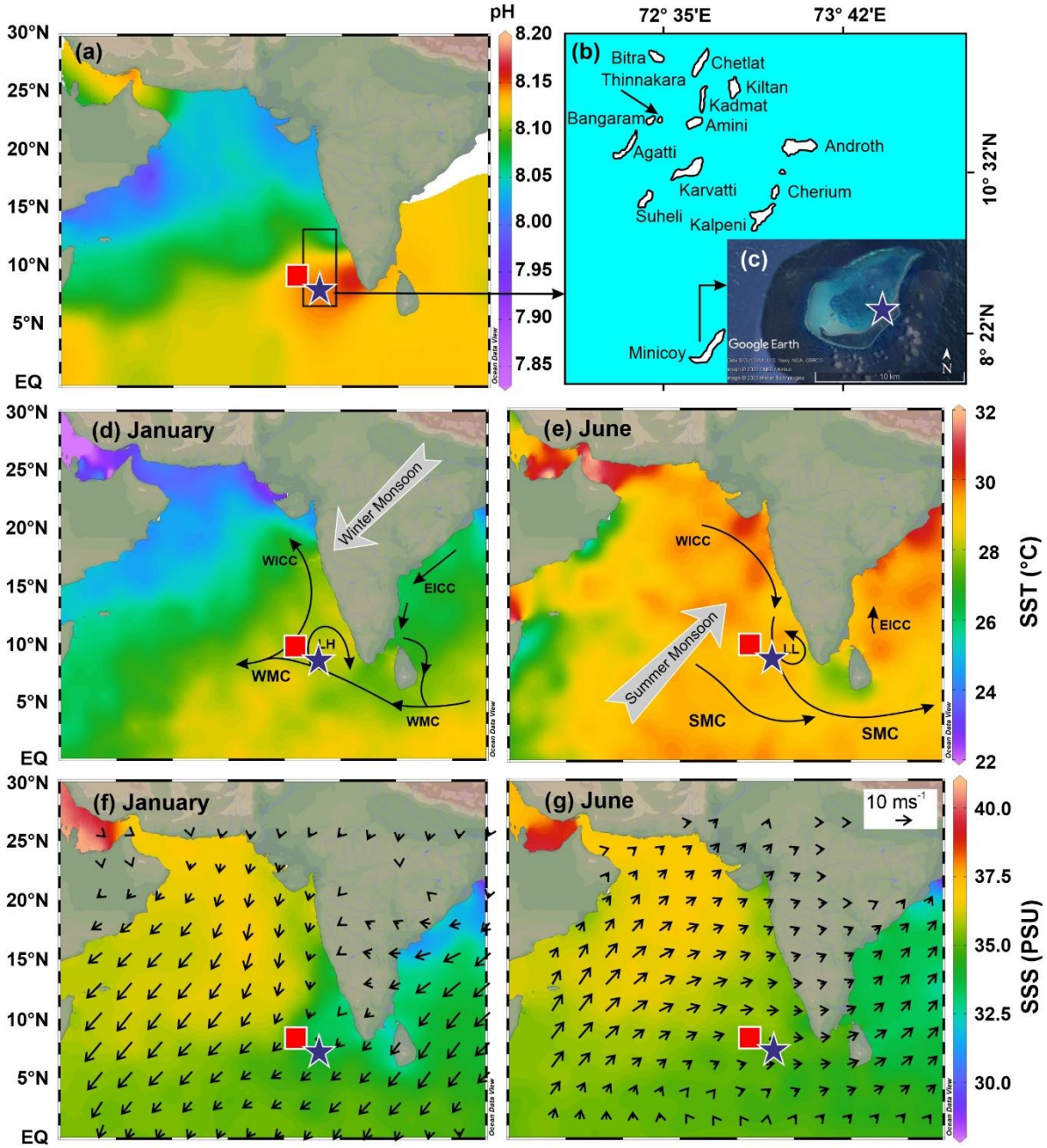


Figure 1 Oceanographic settings and hydrography of the study area. *(a) Surface pH map of the Arabian Sea, (b) Location of islands of Lakshadweep Archipelago, (c) Google earth view of Minicoy Island with the location of the sampling site (blue star). In panels (d) to (g), colour map represents the climatological mean of sea surface temperature (SST) and salinity (SSS) of the Arabian Sea for January and June (World Ocean Atlas 2018). The arrows in panel (f) and (g) represent the climatological mean wind direction and length of the arrow represents the magnitude of wind speed. Blue star represents location of the coral sample and red square represents the location of sediment core, SK221-05 used in this study.*

In this chapter, we have provided the detailed sample processing protocols including foraminifera picking, coral sub-sampling, sample cleaning and dissolution. Further, boron purification from the matrix and analytical protocols has been described in detail. Some of the certified reference materials for the boron isotope ratio ($\delta^{11}B$) were processed throughout the chemistry. The isotope ratio of these standards are well within the reported range. Certified reference material NASS6 was measured after micro-distilled gave an average $\delta^{11}B$ value of 39.964 ‰ ± 0.249 (2σ, n= 8) which is similar to the reported value, 39.81 ‰ ± 0.21 ‰ (2σ, n = 10) (He et al., 2019). Additionally, boric acid standard, European Reference Materials (ERM) AE121 was measured during each analytical session. The long-term average of $\delta^{11}B$ of AE121 is 19.63 ‰ ± 0.20 ‰ (2σ, n = 35) which agrees well with the reported value based on the compilation of five reputed laboratories (19.64‰ ± 0.17‰; Stewart et al., 2021) (Figure 2); thereby, validating the robustness of our analytical method.

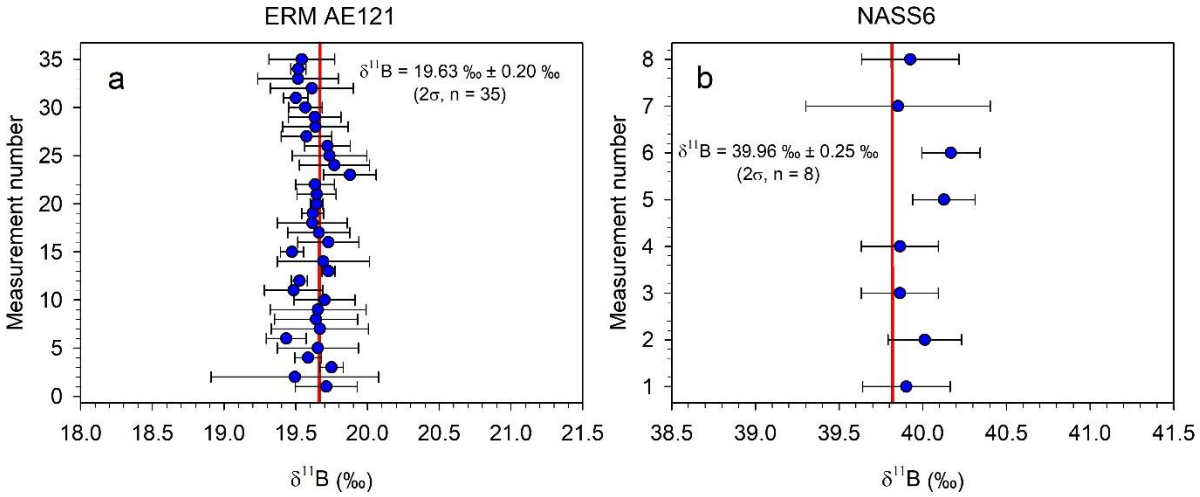


Figure 2 Assessment of the quality of boron isotope analysis. *(a) Measured $\delta^{11}B$ data of ERM AE-121 standard. Red line represents the reported value* (19.64‰ ± 0.17‰; Stewart et al., 2021). *(b) Measured $\delta^{11}B$ data of seawater standard NASS6 after micro-distilled. Red line represents the reported value (*39.81 ± 0.21; He et al., 2019*).*

Chapter 3

Surface pH record (1990–2013) of the Arabian Sea from boron isotopes of Lakshadweep corals—Trend, variability, and control (Published in *JGR: Biogeosciences*)

The main objective of this Chapter was to generate the pH record of the past two decades from the Arabian Sea utilizing the boron isotope and trace-element/Ca ratio of the corals. Further, this chapter also aimed to estimate the ocean acidification trend and to determine the contribution of different forcing factors to the overall pH variability in the Arabian Sea.

Instrument based long-term pH records spanning the last three decades are sparse and restricted to the Pacific and the Atlantic oceans (Bates et al., 2014). However, no such long-term pH record, either instrumental or proxy-based, exists for the Indian Ocean. Therefore, quantifying the ocean acidification rate or determining any long-term trends in the Indian Ocean is challenging. Further, individual forcing factors, both regional and global, that modulate the amplitude and frequency of pH changes in the Indian Ocean are not well understood. In order to fill these research gaps, we have generated a sub-annually resolved pH record (1990–2013) of the eastern Arabian Sea utilising boron isotopic composition ($\delta^{11}B$) of *Porites* corals collected from the Lakshadweep coral reef. Further, using the multi-proxy approach ($\delta^{11}B$ (pH), $\delta^{13}C$ (calcification processes), Sr/Ca and Li/Mg (SST)), we have estimated the contribution of different forcing factors to the overall pH variability in the Arabian Sea. Different proxy records generated in this study and its comparison with ocean climate oscillation index has been provided in Figure 3.

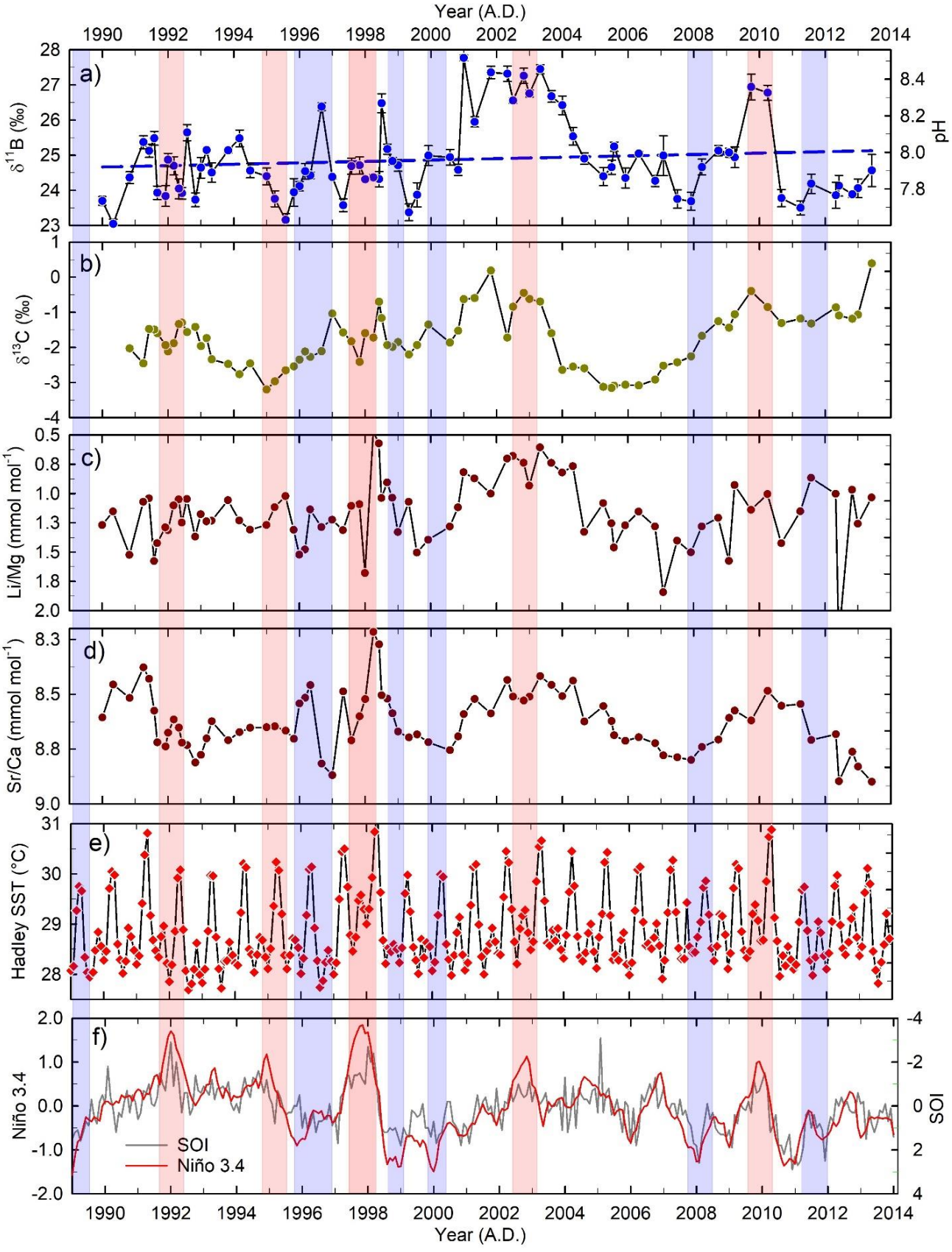

Year (A.D.)
1990
1992
1994
1996
1998
2000
2002
2004
2006
2008
2010
2012
2014
a)
δ11B (‰)
pH
b)
δ13C (‰)
c)
Li/Mg (mmol mol-1)
d)
Sr/Ca (mmol mol-1)
e)
Hadley SST (°C)
f)
Niño 3.4
SOI
SOI
Niño 3.4
Year (A.D.)

Figure 3 Evolution of seawater pH in the Lakshadweep coral reef region, Arabian Sea*. The Panel–a represents $\delta^{11}B$ values (LH-Y axis) of Lakshadweep coral and calculated seawater pH (pH_{SW}) values (RH-Y axis). The dashed line represents the linear regression ($r^2 = 0.015$, n = 79, $p<0.01$) and vertical error bars represent $\pm 2\sigma$ SD uncertainty associated with $\delta^{11}B$ determination. Proxy records of $\delta^{13}C$ (Panel–b) for productivity; Li/Mg ratios (Panel–c) and Sr/Ca ratios (Panel–d) for temperature. The Panel–e represents the Instrumental record of HadlSST at the coral reef site. The Panel–f represents Niño 3.4 time-series (red) and Southern Oscillation Index (SOI) time-series (grey) Red and blue bands represent moderate-strong El-Niño and La-Niña years respectively.*

To summarise this chapter, our coralline $\delta^{11}B$ based sub-annually resolved multi-decadal (1990–2013) pH reconstruction of the western Indian Ocean is characterised by large interannual variability. The predicted long-term declining trend in surface seawater pH from the ~50 ppm rise in atmospheric pCO_2 during the interval of coral growth is obscured by the large internal variability. The observed high amplitude pH variations (extremes) were dominantly modulated by ENSO driven oceanographic processes in the Eastern Arabian Sea. Based on this observation and published numerical simulations of projected increase in ENSO frequency and amplitude, we predict that the future of the massive *Porites* corals in the Arabian Sea is critically dependent on their resilience and adaptability to large bimodal oscillations in pH and SST at sub-annual scale.

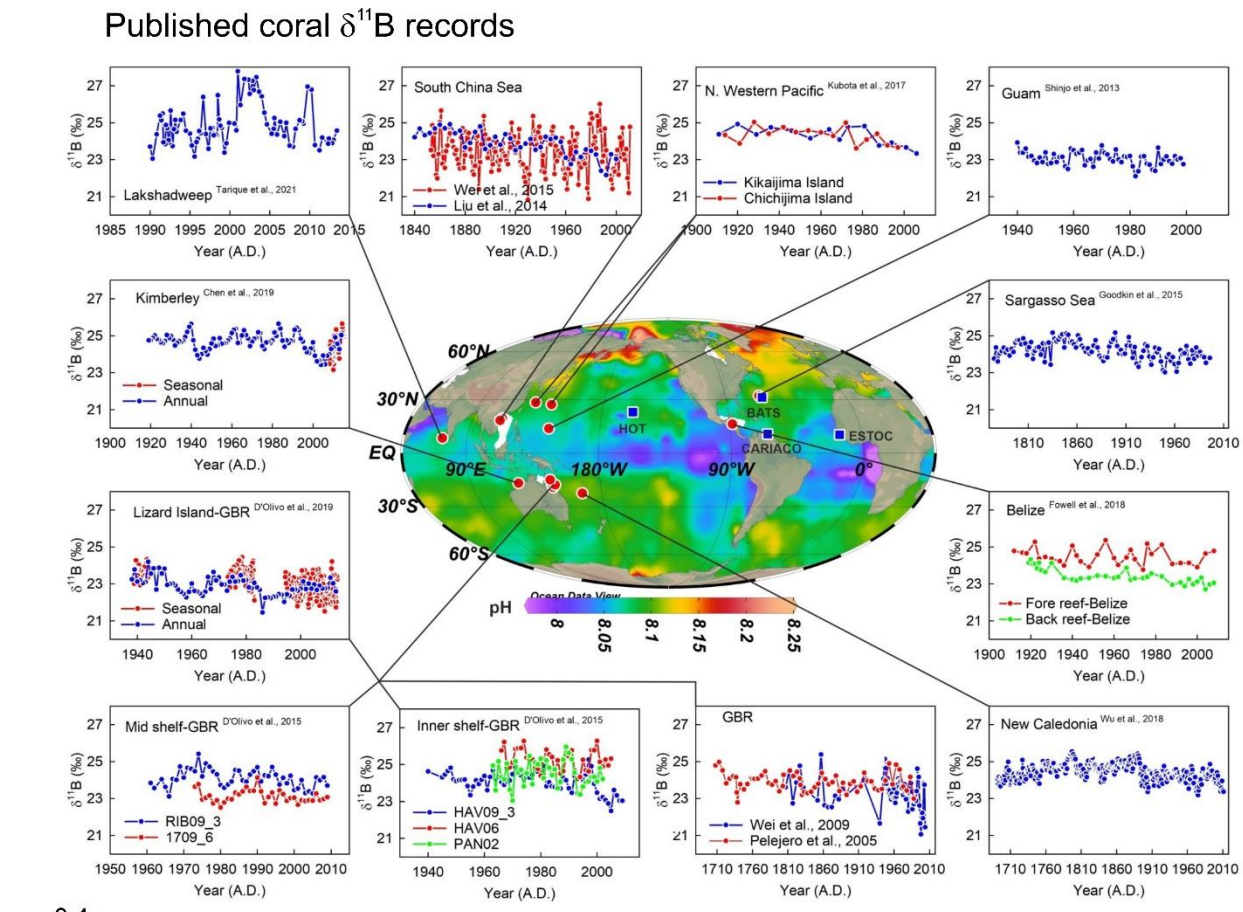
Chapter 4

Global ocean acidification trends from boron isotope ($\delta^{11}B$) records of coral: Role of oceanographic processes and anthropogenic CO_2 forcing

Boron isotope ($\delta^{11}B$) composition of corals has been used to reconstruct pH of ocean for the past several decades to few centuries at resolution ranging from sub-annual to decadal scale. Some of the published studies attempted to decipher the role of anthropogenic CO_2 forcing to recent trends

of ocean acidification. However, such attempts in past were often hindered by limited number of proxy records and knowledge on the relative contribution of different oceanographic factors to the overall pH variability. In this chapter, we have assessed the published coral $\delta^{11}B$ derived pH records and aimed to determine the contribution of different forcing factors to the overall pH variability at sub-annual to decadal scale.

To the best of our knowledge, so far total nineteen (n=19) such records are available from the Pacific (n= 14), Atlantic (n=3) and the Indian oceans (n=2) (Figure 4). Our investigation of $\delta^{11}B$-pH records from the Pacific ocean based on principal component analysis (PCA) reveals that atmospheric CO_2 can explain ~26% of the total pH variability during 1950 – 2009, followed by the ocean-climate oscillations (i.e. ENSO and PDO) driven oceanographic factors, which explains ~17% variability. The remaining large variability (~57%) could be explained by metabolic processes of corals and/or changes in micro-environments, temperature and other physical or biogeochemical processes. Therefore, we highlight the need for detailed investigation in future studies to better understand the exact mechanism and processes/factors that influence the pH variability in coral reef environments. Further, the amplitude of the ENSO driven pH variability shows fivefold increase during 1980 – 2000 AD compared to the previous three decades (1950 – 1980 AD). This observation is consistent with the historical records of global coral bleaching events and therefore underscores the role of ENSO driven environmental stress on coral bleaching. As models predict an increase in ENSO events in future warming scenarios, the "pH extremes" are likely to increase which will relentlessly test the resilience and adaptability" of corals and other calcifiers in global oceans.


Published coral δ11B records
Lakshadweep Tarique et al., 2021
South China Sea
Wei et al., 2015
Liu et al., 2014
N. Western Pacific Kubota et al., 2017
Kikaijima Island
Chichijima Island
Guam Shinjo et al., 2013
Kimberley Chen et al., 2019
Seasonal
Annual
Sargasso Sea Goodkin et al., 2015
Lizard Island-GBR D'Olivo et al., 2019
Seasonal
Annual
Belize Fowell et al., 2018
Fore reef-Belize
Back reef-Belize
Mid shelf-GBR D'Olivo et al., 2015
RIB09_3
1709_6
Inner shelf-GBR D'Olivo et al., 2015
HAV09_3
HAV06
PAN02
GBR
Wei et al., 2009
Pelejero et al., 2005
New Caledonia Wu et al., 2018
δ11B (‰)
Year (A.D.)
60°N
30°N
EQ
30°S
60°S
90°E
180°W
90°W
0°
HOT
BATS
CARIACO
ESTOC
pH
8
8.05
8.1
8.15
8.2
8.25

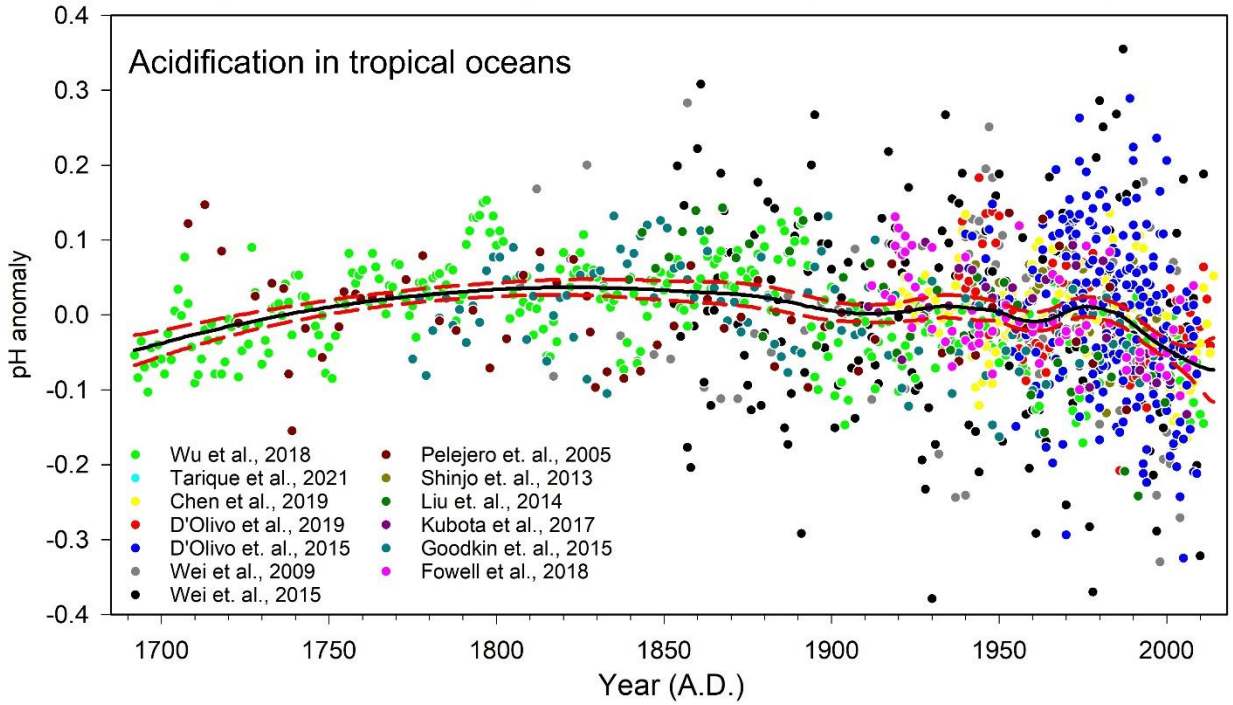

Acidification in tropical oceans
pH anomaly
Year (A.D.)
Wu et al., 2018
Tarique et al., 2021
Chen et al., 2019
D'Olivo et al., 2019
D'Olivo et al., 2015
Wei et al., 2009
Wei et al., 2015
Pelejero et. al., 2005
Shinjo et. al., 2013
Liu et. al., 2014
Kubota et al., 2017
Goodkin et. al., 2015
Fowell et al., 2018

Figure 4 Published boron isotope record of corals: *Location of the published coral boron isotope records is shown by red circle. Each panel represent the $\delta^{11}B$ time-series at the corresponding location. Blue square represents the location of established in-situ pH time-series stations i.e. HOT in the Pacific and BATS, CARIACO and ESTOC in the Atlantic. Colour band represent pH of the surface ocean.*

Chapter 5

Quantifying the CO_2 source/sink variability in tropical ocean during the last glacial cycle

Changes in the Glacial–interglacial (GIG) atmospheric CO_2 are generally attributed to changes in seawater carbon chemistry resulting from change in the ocean's biogeochemistry and circulation (Sigman et al., 2010). However, the physical, biological and chemical variables that control ocean–atmosphere CO_2 exchange during glacial–interglacial cycles have not been well understood. The Indian Ocean has been an important but poorly studied ocean basin. The modern Arabian Sea, Western Indian Ocean is the second largest CO_2 source region after Eastern Equatorial Pacific (Sarma, 2003). Therefore, tracing the CO_2 source/sink variability of the Arabian in the past is of great importance in understanding the mechanism of glacial-interglacial CO_2 variability. So far only two $\delta^{11}B$ derived pH/pCO_2 records are available from the Arabian Sea in the Indian Ocean (Naik et al., 2015; Palmer et al., 2010) (Figure 5), both of these records cover the time period up to the last glacial maxima (LGM). One of these records represents the Northern Arabian Sea (Palmer et al., 2010), whereas the other record is from the eastern Arabian Sea (Naik et al., 2015). Records of the Naik et al. (2015) is located near the coast and influenced by monsoonal runoff and act as a sink of CO_2 albeit most of the Arabian Sea acts as a strong source and hence does not represent the overall Arabian Sea surface CO_2 conditions. Secondly, the resolution and time span of these records are not enough to resolve several important climate transitions such as the Heinrich-Stadial (HS), Younger Dryas (YD), last glacial maxima (LGM) and other events during the last glacial cycle.

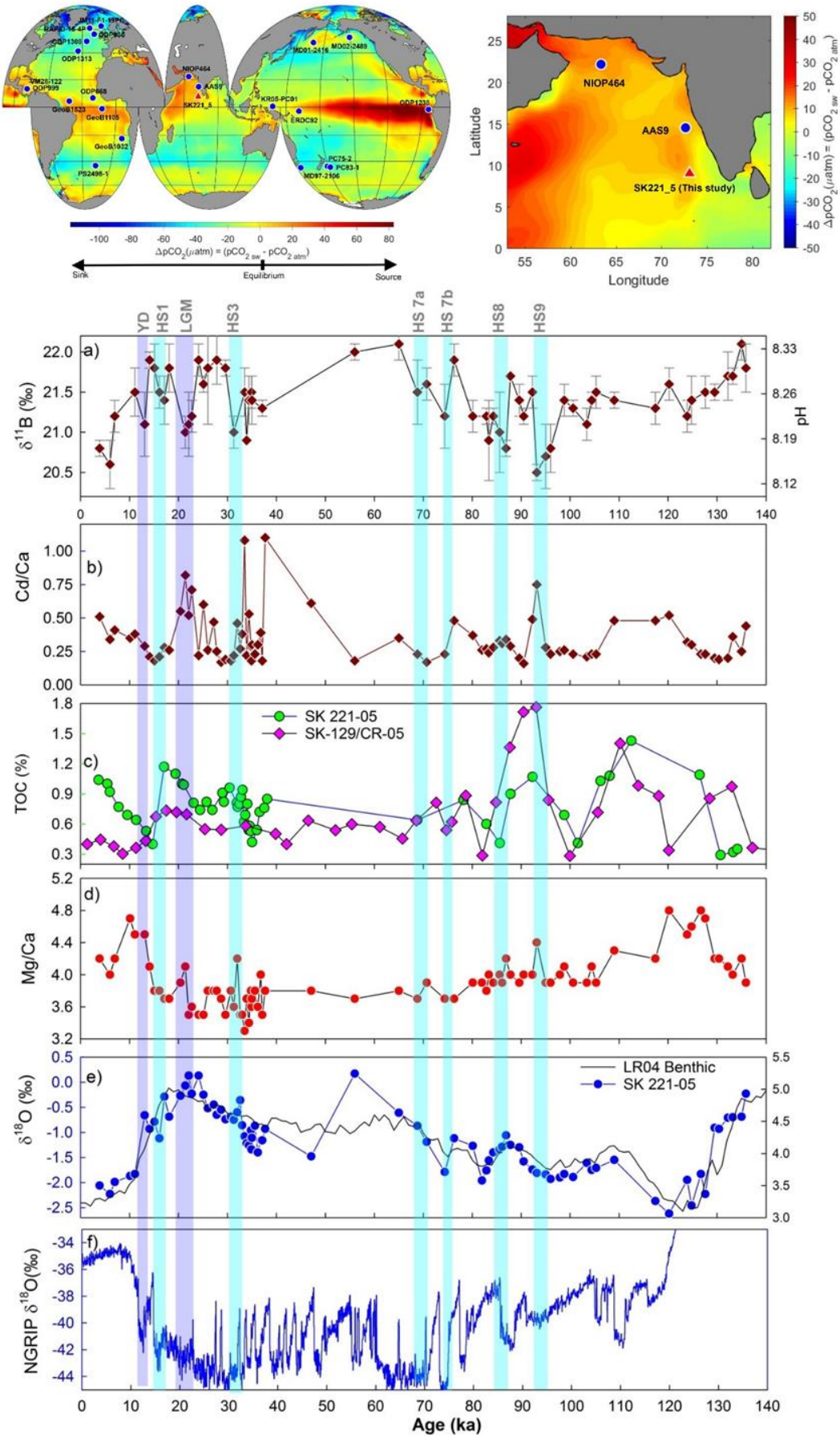

NIOP464
AAS9
SK221_5
KR05-PC01
ERDC92
MD01-2416
MD02-2489
ODP1238
PC75-2
PC83-3
MD97-2106
ODP1313
ODP999
ODP668
GeoB1523
GeoB1105
GeoB1032
PS2498-1
-100 -80 -60 -40 -20 0 20 40 60 80
ΔpCO2(μatm) = (pCO2 sw - pCO2 atm)
Sink
Equilibrium
Source
NIOP464
AAS9
SK221_5 (This study)
Latitude
Longitude
ΔpCO2(μatm) = (pCO2 sw - pCO2 atm)
YD
HS1
LGM
HS3
HS 7a
HS 7b
HS8
HS9
a)
δ11B (‰)
pH
b)
Cd/Ca
c)
SK 221-05
SK-129/CR-05
TOC (%)
d)
Mg/Ca
e)
LR04 Benthic
SK 221-05
δ18O (‰)
f)
NGRIP δ18O(‰)
Age (ka)

Figure 5 Global surface ΔpCO_2 map and proxy records from the Arabian Sea. *Top left panel - Global surface ΔpCO_2 map. Blue circles represent the location of the published $\delta^{11}B$ records of the planktic foraminifera whereas the red star represents the location of the present study. Top right panel - Surface ΔpCO_2 map of the Arabian Sea. Bottom panel (a) The $\delta^{11}B$ record and its corresponding pH, (b) record of Cd/Ca ratio, a proxy for nutrient (PO_4^-), (c) record of TOC % record from the core SK221-05 (present study) and another record (SK-129/CR-05) from the proximity, (d) Mg/Ca record as proxy for sea surface temperature (SST), (e) record of SK221-05 G. ruber $\delta^{18}O$ is plotted along with the LR04 benthic $\delta^{18}O$ stack. (f) North Greenland Ice Core Project (NGRIP) $\delta^{18}O$ record.*

Therefore, in order to have a holistic picture of the tropical oceans and to assess the contribution of tropical oceans to the atmospheric CO_2 evolution in the last glacial-interglacial cycle, it is imperative to have more records from the Arabian Sea. In order to fill this data gap in the Indian Ocean. Here, we provide a high-resolution record of pH/pCO_2 from the eastern Arabian derived from boron isotope of planktic foraminifera (*Globigerinoides ruber*). Further, we have compiled the published planktic foraminifera $\delta^{11}B$ derived pCO_2 records (Figure 6).

The surface pCO_2 in the Arabian Sea is influenced by the multiple factors such as seasonal variation in the wind pattern, surface circulation, upwelling and biological productivity. Thus, the source and sink variability in this region over the last glacial cycle are challenging to interpret. Here, we present new Mg/Ca, $\delta^{18}O$, Cd/Ca and $\delta^{11}B$ data from Eastern Arabian Sea covering the last 135 kiloyears. These multi-proxy data of the same core allow us to investigate the mechanisms that regulate change in seawater CO_2 during GIG intervals. Our results from Eastern Arabian Sea sediment core, SK221-5 show that pCO_{2SW} varied between ~160 and 300 μatm over the past 130 kyr and this region acted as a source for atmospheric CO_2 during the Last Glacial Maximum. We demonstrate that the Eastern Arabian Sea exhibits considerable variability in terms of its carbonate chemistry and CO_2 source/sink through the entirety of the last glacial cycle. CO_2 source capacity of the Arabian Sea was amplified during the cold events such as Last Glacial Maxima (LGM), Heinrich Events (HE) and Younger Dryas (YD). This could be due to northward invasion of AAIW in the Arabian Sea coupled with enhanced upwelling during the cold periods.

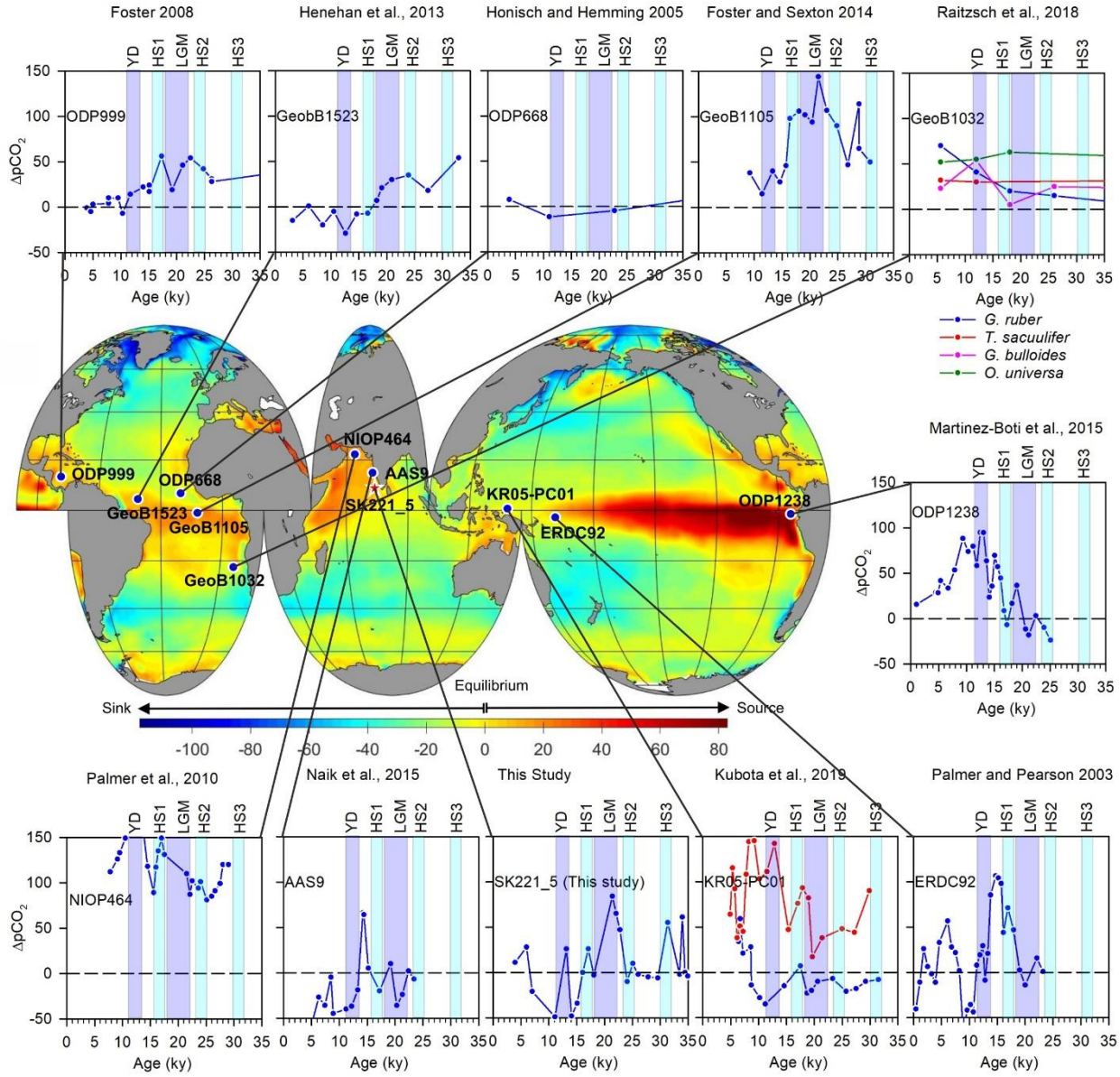


Figure 6 Global Sea surface ΔpCO_2 map and proxy derived ΔpCO_2 timeseries. *(a) Global surface ΔpCO_2 map, where red colour represents the areas of modern CO_2 source to the atmosphere and blue colour show CO_2 sink regions. The ΔpCO_2 map was generated by subtracting the atmospheric pCO_2 values from the seawater pCO_2 values. Blue circles represent the location of the published boron isotope based ΔpCO_2 records of the planktic foraminifera whereas the red star represents the location of the present study. Here ΔpCO_2 records only from the tropics have been plotted. Each panel represents the ΔpCO_2 at corresponding location.*

Chapter 6

Summary and Conclusions

Some of the major observations and findings of this thesis has been summarised below:

- Our corals $\delta^{11}B$ derived sub-annual resolution pH record shows that the long-term acidification trend in the Arabian Sea is obscured by large internal variability.
- Physical processes modulated by ENSO dominantly controls large inter-annual pH variability in the Arabian Sea.
- Compilation of the published coral $\delta^{11}B$/pH records from the Pacific, Atlantic and Indian Ocean show a discernible decreasing trend since the industrial era, ~1850 AD.
- Other than atmospheric CO_2 concentration, the pH variability in the Pacific Ocean is dominantly controlled by ENSO, whereas, in the Atlantic, it is modulated by NAO and AMO driven oceanographic factors/processes.
- In the Pacific Ocean, atmospheric CO_2 together with ocean-climate oscillations driven oceanographic factors explain ~37% of the total pH variability.
- Our foraminifera $\delta^{11}B$ derived pCO_2 record shows that the Arabian Sea acted as a source of CO_2 to the atmosphere for most of the time during the last glacial cycle.
- CO_2 source capacity of the Arabian Sea was amplified during the cold events such as Last Glacial Maxima (LGM), Heinrich Events (HE) and Younger Dryas (YD). This could be due to northward invasion of AAIW in the Arabian Sea coupled with enhanced upwelling during the cold periods.

List of Publications

Published articles

Mohd Tarique, Waliur Rahaman, Fousiya A.A., N. Lathika., Meloth Thamban, Hema Achyuthan, Sambuddha Misra, 2021, Surface pH record (1990–2013) of the Arabian Sea from boron isotopes of Lakshadweep corals—trend, variability, and control. ***Journal of Geophysical Research: Biogeosciences***, 126, e2020JG006122, DOI: 10.1029/2020JG006122

N. Lathika, Waliur Rahaman, **Mohd Tarique**, Naveen Gandhi, Avinash Kumar, Meloth Thamban, 2021, "Deep water circulation in the Arabian Sea during the last glacial cycle: Implications for paleo-redox condition, carbon sink and atmospheric CO_2 variability", ***Quaternary Science Reviews***, 257: 106853, DOI: 10.1016/j.quascirev.2021.106853

Waliur Rahaman, Lukas Smik, Deniz Köseoğlu, N Lathika, **Mohd Tarique**, Meloth Thamban, Alan Haywood, Simon T Belt, Jochen Knies, 2020. "Reduced Arctic sea ice extent during the mid-Pliocene Warm Period concurrent with increased Atlantic-climate regime", ***Earth and Planetary Science Letters***, 550: 116535. DOI: 10.1016/j.epsl.2020.116535

Manuscript under review

Mohd Tarique and Waliur Rahaman, 2021, Recent ocean acidification trends from boron isotope record of corals: Role of oceanographic processes and anthropogenic CO_2 forcing, ***Geoscience Frontiers*** (Manuscript# GSF-S-21-00742)

Manuscript under preparation

Mohd Tarique, Waliur Rahaman, N. Lathika, Sambuddha Misra, 2021, Strong CO_2 outgassing from the tropical oceans during colder periods of the last glacial cycle

Papers Presented at Seminar/Conference

Mohd Tarique and Waliur Rahaman "Assessment of boron isotope derived pH record from Pacific and Atlantic Ocean Corals: Oceanographic factors versus Anthropogenic CO_2 forcing", National Conference on Polar Sciences, 16-17th May 2017, at ***National Conference on Polar Sciences*** held at Goa.

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Signature of the Student Signature of the Supervisor

Date…… Date…….